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Approved For Release 1999/09/24 : CIA-RDP83-00423R002000130009-6

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THE INFLUENCE OF ELASTIC SHEAR STRAINS ON  
THE CONDUCTIVITY AND THERMO-ELECTRIC  
FORCE OF CUBIC METALS

BY  
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GEPUBLICEERD IN: PHYSICA 18, 587-596, AUGUSTUS/SEPTEMBER 1952

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Physica XVIII, no 8-9

Augustus-September 1952

## THE INFLUENCE OF ELASTIC SHEAR STRAINS ON THE CONDUCTIVITY AND THERMO-ELECTRIC FORCE OF CUBIC METALS

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### Synopsis

Elastic shear strains cause a change in the shape of the Fermi surface of metals. The influence that such a change has upon the electrical conductivity and the thermo e.m.f. has been calculated for monovalent f.c.c. metals, and the results of it have been compared with the experimental values for Cu, Ag and Au. The conclusion can be drawn that the Fermi surface of gold touches the Brillouin zone boundary, whereas that of copper does not, whilst silver is an intermediate case. The theoretical results account also qualitatively for the experimental values of the trivalent Al.

1. *Introduction.* Even in cubic monovalent metals the Fermi surface in wavevector space for the conduction electrons is not spherical. For sodium it is generally assumed that the deviations from the spherical shape are negligible, but for the noble metals this is probably not the case. The electrical conductivity is isotropic for cubic crystals, but if it could be made anisotropic by artificial means, it might be possible to obtain information about the anisotropy of the Fermi surface. This can indeed be done by applying elastic shear strain.

The linear dependence of the resistance on the strain can be described by a fourth-order tensor, since it relates two second order tensors. In the case where the coördinate axes coincide with the cubic axes of the crystal one obtains:

$$\begin{aligned}\Delta \varrho_{xx}/\varrho_0 &= -\varrho_0 \Delta \sigma_{xx} = \alpha_{11} \varepsilon_{xx} + \alpha_{12} (\varepsilon_{yy} + \varepsilon_{zz}) \\ \Delta \varrho_{xy}/\varrho_0 &= -\varrho_0 \Delta \sigma_{xy} = 2\alpha_{44} \varepsilon_{xy}\end{aligned}\quad (1)$$

and similar expressions for the other directions. Here  $\varrho_0$  is the resistance in the undeformed state, and  $\sigma_{ij}$  the conductivity. The

constants  $a_{ij}$  are analogous to the elastic constants, and therefore the same abbreviations are used. The three combinations of interest are  $a_{11} + 2a_{12}$ ,  $a_{11} - a_{12}$  and  $2a_{44}$ . The first quantity gives the change under hydrostatic pressure and the last two the changes under shear strains, leaving the volume constant. For isotropic materials the last two quantities are equal:  $a_{11} - a_{12} = 2a_{44}$ . The theory for  $a_{11} + 2a_{12}$  has been given by Mott<sup>1)</sup>. Under hydrostatic pressure the elastic constants increase due to the anharmonic term in the potential energy, which also gives rise to the thermal expansion. Thus the amplitude of the lattice vibrations is decreased, giving a smaller resistance and  $a_{11} + 2a_{12} > 0$ . The theory accounts satisfactorily for the experimental values. Here we shall be concerned only with  $a_{11} - a_{12}$  and  $2a_{44}$ .

Another property strongly depending on the conduction electrons near the Fermi surface is the absolute thermo-electric power, being also a second-order tensor and thus isotropic for cubic crystals. The changes under the application of strain are given by equations analogous to (1):

$$\begin{aligned} \Delta e_{xx} &= \beta_{11}\epsilon_{xx} + \beta_{12}(\epsilon_{yy} + \epsilon_{zz}) \\ \Delta e_{xy} &= 2\beta_{44}\epsilon_{xy}, \end{aligned} \quad (2)$$

$\beta_{ij}$  being the components of a fourth-order tensor, giving the change in the thermo e.m.f.  $e_{ij}$ . In this case the  $\beta_{ij}$  are not dimensionless quantities, as in (1). We did not divide by the values for zero strain, as this quantity may be either positive or negative, as well as zero. The element  $\beta_{44}$  gives rise to effects as observed by P e r r i e r<sup>2)</sup> (transverse currents in a twisted tube in which a longitudinal temperature gradient exists), though the mechanism is quite different for ferromagnetic metals. It will be shown below that the  $\beta$ 's are closely related to the  $a$ 's, and that both can give us qualitative information as to the shape of the Fermi surface.

2. *Derivation of basic formulae.* The electrical current density is given by

$$j_i = -2e/\hbar(2\pi)^3 \int \int \int_{-\infty}^{+\infty} \partial E(\mathbf{k})/\partial k_i f(\mathbf{k}) dk_x dk_y dk_z, \quad (3)$$

where  $-e$  is the electronic charge,  $f(\mathbf{k})$  is the probability that an electron is in the state characterized by the wave vector  $\mathbf{k}$ , and  $E(\mathbf{k})$  the energy of that state. In the presence of an electrical field strength  $\mathbf{F}$  and a temperature gradient, for  $f(\mathbf{k})$  we can take

$$f(\mathbf{k}) = f_0(E) + df_0/dE (e\mathbf{F} + (E - \zeta)/T, \nabla T + \nabla\zeta) \cdot \boldsymbol{\lambda}(\mathbf{k}), \quad (4)$$

with  $\lambda(\mathbf{k})$  satisfying the integral equation

$$\nabla_{\mathbf{k}} E(\mathbf{k}) = \int_{\text{Fermi}} \{\lambda(\mathbf{k}) - \lambda(\mathbf{k}')\} P(\mathbf{k}\mathbf{k}') (\partial E / \partial k_n)^{-1} dS' \quad (5)$$

$f_0(E)$  is the Fermi-Dirac function  $f_0(E) = (\exp. (E - \zeta)/kT + 1)^{-1}$ , and  $\zeta$  the thermodynamic potential of the electrons.  $P(\mathbf{k}\mathbf{k}')$  is, apart from a numerical factor, the transition probability. In (5) it is assumed that the energy is conserved during a transition ( $T > 0_D$ ), the integration has to be taken over the Fermi surface in  $\mathbf{k}'$  space. The introduction of the vector field  $\lambda(\mathbf{k})$  is in general not equivalent to that of a relaxation time  $\tau(\mathbf{k})$ . The use of this last quantity means that  $\lambda(\mathbf{k}) \equiv \tau(\mathbf{k}) (1/\hbar) \nabla_{\mathbf{k}} E(\mathbf{k})$ , but for the general case of non-spherical Fermi surface and a transition probability which does not depend solely upon the angle between  $\mathbf{k}$  and  $\mathbf{k}'$ ,  $\lambda(\mathbf{k})$  will not always be parallel to  $\nabla_{\mathbf{k}} E(\mathbf{k})$ , and the use of  $\tau(\mathbf{k})$  is not correct.

The conductivity is given by

$$\sigma_{ij} = 2e^2/\hbar(2\pi)^3 \cdot \int_{\text{Fermi}} \cos(n, i) \lambda_j(\mathbf{k}) dS \quad (6)$$

where for  $df_0/dE$  has been taken  $-\delta(E - \zeta)$ , this being valid for not too high temperatures. After multiplying (3) by the resistance tensor  $\varrho_{ij}$  we find

$$F_i = j_i \varrho_{ij} + \frac{1}{eT} \cdot \frac{\partial T}{\partial x_k} \varrho_{ij} \int_0^\infty (E - \zeta) \frac{df_0}{dE} \cdot S_{ik}(E) dE - \frac{1}{e} \frac{\partial \zeta}{\partial x_j} \quad (7)$$

with

$$S_{ij}(E) = 2e^2/\hbar^2(2\pi)^3 \cdot \int \cos(n, i) \lambda_j(\mathbf{k}) dS \quad (8)$$

where the integration in (8) has to be taken over the surface with constant energy  $E$ . Comparing (8) with (6) it follows that  $\sigma_{ij} = S_{ij}(\zeta)$ . Making use of the wellknown series expansion for integrals containing  $df_0/dE$ , one finds for the absolute thermo-electric force tensor

$$e_{ij} = -\pi^2 k^2 T / 3e \cdot \varrho_{ik} (dS_{kj}(E)/dE)_{E=\zeta} \quad (9)$$

In order to determine the change of (9) under elastic strain we make use of the generalized equations (1):

$$\varrho_{ij} = \varrho_0(\delta_{ij} + \alpha_{ijkl} \varepsilon_{kl}) \quad S_{ij}(E) = S_0(E) (\delta_{ij} - \alpha_{ijkl}(E) \varepsilon_{kl})$$

and by means of (9) find

$$\partial e_{ij} / \partial \varepsilon_{kl} = \beta_{ijkl} = \pi^2 k^2 T / 3e (d\alpha_{ijkl}(E)/dE)_{E=\zeta} \quad (10)$$

3. *The change of the Fermi surface under shear strain.* One of the most characteristic features of the influence of elastic shear strain

is the change in the shape of the non-spherical parts of the Fermi surface. This is most easily seen in the one-dimensional case (Fig. 1). A strain  $\epsilon_{kl}$  of the specimen corresponds to a strain  $-\epsilon_{kl}$  of the  $k$  space. We shall use the nearly-free-electron approximation. The energy gap is  $2V_g$ ,  $V_g$  being the Fourier component of the periodic potential corresponding to the repetency vector  $g$  of the zone boundary. Moreover we assume that electrons near one particular energy gap are only affected by that gap. Strains which leave the volume constant do not alter  $\zeta$ . In fig. 1 there is an extension

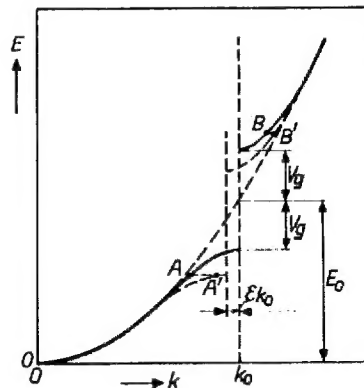


Fig. 1. One-dimensional energy curve for nearly free electrons. The change in it due to strain is dashed.

( $\epsilon_{xx} = \epsilon > 0$ ) in the drawn direction, and compression in directions perpendicular to it ( $\epsilon_{yy} = \epsilon_{zz} = -\epsilon/2$ ). The change in  $V_g$  is immaterial, since  $V_g$  is small compared with  $E_0$  in this approximation. Moreover it is constant for the "deformierbare" potential of Bloch a.o., as used in the theory of the interactions between the lattice vibrations and the conduction electrons. Bardén<sup>3)</sup> has shown that this is a good approximation for monovalent metals. But also for the rigid ions of Nordheim the change in the energy gap will be small as compared with that of  $E_0$ . Consequently we shall ignore it. The point  $A$  in the first Brillouin zone goes over into  $A'$ , much nearer to the boundary and having a smaller slope  $dE/dk$ . We can say that the electron is less free than in  $A$ , as a consequence of the increased lattice parameter in that direction. This fact increases the resistance in this direction. Since it corresponds to  $a_{11} - a_{12}$  or  $2a_{44}$ , these quantities should be positive. In the case of

$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon > 0$  the same picture of fig. 1 applies, but then  $\zeta$  is no longer constant, and  $A'$  will be lower than  $A$ . These effects are therefore more important for purely shape changes than for volume changes. It is easily seen that much the same applies for holes within the first Brillouin zone. The contrary is true in a point  $B$  of the second Brillouin zone, where  $B'$  is nearer to the free electron parabola than  $B$ , leading to negative  $\alpha_{11} - \alpha_{12}$  or  $2\alpha_{44}$ . It may be stressed that for free electrons these effects vanish, and that the largest changes may be expected for points very near a Brillouin zone boundary, which for f.c.c. monovalent metals lie in the (111) directions. Extension  $\varepsilon$  therein and equal compression perpendicular to it, leaving the volume constant, gives in the (111) direction a relative change in resistance of  $2\alpha_{44}\varepsilon$ . From these very simple considerations one should expect for the monovalent f.c.c. metals  $2\alpha_{44} > \alpha_{11} - \alpha_{12} > 0$ , and for those of higher valency with electrons in the second zone  $2\alpha_{44} < \alpha_{11} - \alpha_{12} < 0$ . These qualitative results are now in striking agreement with the experimental values published by D r u y v e s t e i n <sup>4)</sup> for the noble metals Cu, Ag and Au and for the trivalent Al. Moreover it is seen that  $\alpha$  will be larger, the higher the F e r m i surface as long as it does not touch the zone boundary. According to (10) then  $\beta_{11} - \beta_{12}$  or  $2\beta_{44}$  will be positive. Following the same reasoning as before we get  $2\beta_{44} > \beta_{11} - \beta_{12} > 0$ . But the contrary applies for holes within the first Brillouin zone, since these holes will become more "free" for higher energy. It will be shown that this is also in accordance with the experiments, and therefore it seems worth-while to investigate this aspect of the problem more quantitatively.

From (6) and (5) it follows that

$$\Delta\sigma_{ii} = 2e^2/\hbar^2 \cdot (2\pi)^3 \Delta \int \cos(n, i) g_i(\mathbf{k}) dS \quad (11)$$

$$\Delta \frac{\partial E(\mathbf{k})}{\partial k_i} = \Delta \lambda_i(\mathbf{k}) \cdot \int P(\mathbf{k}\mathbf{k}') \left( \frac{\partial E}{\partial k'_n} \right)^{-1} dS' + \lambda_i(\mathbf{k}) \cdot \Delta \int P(\mathbf{k}\mathbf{k}') \left( \frac{\partial E}{\partial k'_n} \right)^{-1} dS' - \Delta \int \lambda_i(\mathbf{k}') P(\mathbf{k}\mathbf{k}') \left( \frac{\partial E}{\partial k'_n} \right)^{-1} dS' \quad (12)$$

For shape changes under constant volume the first two terms of (12) are the most interesting. For an isotropic transition probability the last two terms should vanish. They contain the influence of the change in the lattice vibrations and depend also on the change in

the properties of quantities at the Fermi surfaces, but to a lesser degree than the first two terms of (12), because of the integration. Also in the present case of non-isotropic  $P(\mathbf{k}\mathbf{k}')$  we shall ignore the last two integrals in (12). In the same approximation we shall assume the coefficient of  $\Delta\lambda_i(\mathbf{k})$ , i.e.  $\int P(\mathbf{k}\mathbf{k}') (\partial E / \partial k'_i)^{-1} dS'$  to be isotropic. In general  $\lambda(\mathbf{k})$  will not be parallel to  $\nabla_{\mathbf{k}} E(\mathbf{k})$ , but in this approximation the changes in it are parallel. We now get for the relative change in the conductivity

$$\frac{\Delta\sigma_{ii}}{6} = -\frac{\Delta\rho_{ii}}{\rho} = \left( \int \frac{\partial E}{\partial k_i} dS_i \right)^{-1} \cdot \Delta \int \frac{\partial E}{\partial k_i} dS_i,$$

from which also the change in thermo e.m.f. can be derived. For zero strain we shall take the free electron value. Calculations are then made for the f.c.c. monovalent metals by dividing the  $\mathbf{k}$  space into pyramids formed by the intersections of the discontinuity planes and with their apex in the origin as indicated in fig. 2, and after-

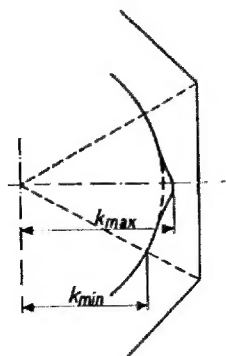


Fig. 2. The partition of the Brillouin zone into pyramids.

wards replacing these pyramids by cones of revolution having the same solid angle. The points within one cone are assumed to be affected only by their own energy gap. For the f.c.c. lattice there are 8 (111) discontinuity planes and 6 (100) ones, having energy gaps  $2V_{111}$  and  $2V_{100}$  respectively. The energy in this approximation is given by

$$E(\mathbf{k}) = \frac{\hbar^2 (\mathbf{k} - \mathbf{g})^2 + (\mathbf{k})^2}{2m} \pm \sqrt{\left\{ \frac{\hbar^2 (\mathbf{k} - \mathbf{g})^2 - (\mathbf{k})^2}{2m} \right\}^2 + |V_{\mathbf{g}}|^2}$$

and we find for  $\int \partial E / \partial k_i dS_i$  in the direction of the axis of the cone ( $\xi$ ) and perpendicular to it ( $\eta$ ) respectively

$$\sigma_{\xi\xi} = \frac{2\pi m}{\hbar^2} \int_{k_{\min}}^{k_{\max}} \left( \frac{\partial E}{\partial k_\xi} \right)^2 dk_\xi \quad \text{and} \quad \sigma_{\eta\eta} = \frac{\pi \hbar^2}{m} \int_{k_{\min}}^{k_{\max}} k_\eta^2 dk_\xi$$

The contributions for each cone towards the changes in conductivity and thermo e.m.f. due to a shape change could now be calculated and then summed up. The results are plotted in fig. 3 for  $\alpha_{11} - \alpha_{12}$

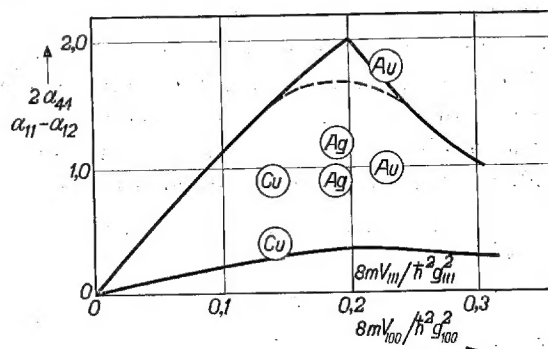


Fig. 3. The calculated curve for  $\alpha_{11} - \alpha_{12}$  (lower curve) as  $2\alpha_{44}$  (upper curve) as a function of  $8 mV_{100} / \hbar^2 g_{100}^2$  and  $8 mV_{111} / \hbar^2 g_{111}^2$  respectively. The experimental points are indicated.

and  $2\alpha_{44}$  and in fig. 4 for  $\beta_{||} - \beta_{\perp}$  ( $T = 300^\circ\text{K}$ ). For the change in the thermo-electric force only experimental values for polycrystalline samples were known, and therefore we have to average. The result is  $\beta_{||} - \beta_{\perp} = 0,4 (\beta_{11} - \beta_{12}) + 1,2 \beta_{44}$ . The calculated positive values are practically only due to  $\beta_{44}$  since  $\beta_{11} - \beta_{12}$  is very small or negative. In plotting the figures it has been assumed for simplicity that  $V_{111}/g_{111}^2 = V_{100}/g_{100}^2$  but this is immaterial.

4. Discussion and conclusions. In the table the experimental values of Druvesteyn<sup>4)</sup> are given for the resistance effect and those reviewed by Borelius<sup>5)</sup> for the thermo-electric force

	$\alpha_{11}$	$\alpha_{12}$	$2\alpha_{44}$	$\alpha_{11} - \alpha_{12}$	$\alpha_{11} + 2\alpha_{12}$	$\beta_{  } \cdot 10^8 \text{V}/^\circ\text{K}$	$\beta_{\perp} \cdot 10^8 \text{V}/^\circ\text{K}$	$(\beta_{  } - \beta_{\perp}) \cdot 10^8 \text{V}/^\circ\text{K}$	$(\beta_{  } + 2\beta_{\perp}) \cdot 10^8 \text{V}/^\circ\text{K}$	$E \cdot 10^{-12} \text{dyne/cm}^2$	$\mu$
Cu	2.6	2.2	0.9	0.4	7.0	9.5	2.1	7.4	13.6	1.24	0.35
Ag	4.0	3.1	1.2	0.9	10.2	12.9	6.7	6.2	26.4	0.79	0.37
Au	6.3	5.3	1.8	1.0	16.9	8.0	7.4	0.6	22.9	0.78	0.42
Al	2.5	3.5	-2.0	-1.0	9.5	-12.9	-10.7	-2.2	-34.2	0.70	0.34



value after expressing them in terms of strains, using the elastic constants reviewed by Druyvesteyn<sup>6</sup>).

Theoretically  $2a_{44}$  rises as far as the point where the Fermi surface touches the Brillouin zone boundary ( $8mV_{111}/\hbar^2 g_{111}^2 = 0.2$ ), after which it drops again. The maximum value is about 2.0. It cannot yet be concluded whether the Fermi surface touches the zone boundary or not. For  $2\beta_{44}$  there is a sharp discontinuity at the critical point. From the lower side of  $8mV_{111}/\hbar^2 g_{111}^2$  the term goes to infinity, but for values exceeding 0.2, it is finite and negative; negative because of the occurrence of holes. The calculated discontinuity will not occur because of lattice imperfections and the thermal vibrations of the lattice. At room temperature the mean amplitude of the ions is 3% of their distance, so the unsharpness in the energy is about 6%. The estimated curve is dashed.

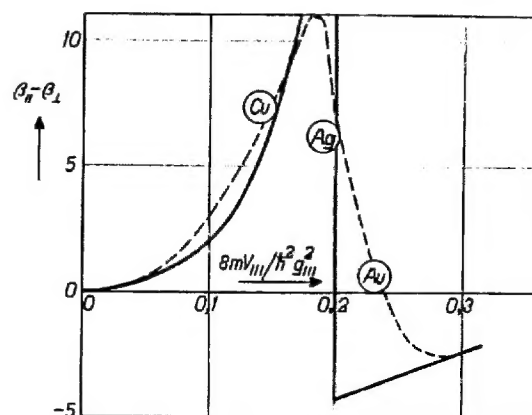


Fig. 4. The calculated curve for  $\beta_{||} - \beta_{\perp}$  as a function of  $8mV_{111}/\hbar^2 g_{111}^2$ . The experimental points are indicated.

The approximations made to arrive at the values of  $\alpha_{11} - \alpha_{12}$  and  $2a_{44}$  are very drastic, e.g. the influence of the change in the lattice vibrations is ignored. The other ignored influences on the transition probability etc. are of minor importance. It is well known that for the noble metals Cu, Ag and Au the elastic constants are not very much influenced by the Fermi energy. The ions may be considered as being hard spheres touching each other. Therefore the change in lattice vibrations does not play a part in determining the  $\beta$ 's and so for these our calculations apply much better. The thermoelectric

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## INFLUENCE OF SHEAR ON CONDUCTIVITY OF CUBIC METALS 595

power has been calculated by Mott and Jones<sup>7)</sup>. They showed that it is negative for nearly free electrons, and positive for holes. It is also positive if the relaxation time decreases strongly with increasing  $\zeta$ , as for electrons near a zone boundary. This last effect may explain the positive thermo e.m.f. of the noble metals. The effect of all-sided extension ( $\beta_{11} + 2\beta_{12}$ ) is now to make the electrons less free, so the thermo e.m.f. will increase and  $\beta_{11} + 2\beta_{12} > 0$ , in agreement with the experiments. Since the larger part of  $\alpha_{11} + 2\alpha_{12}$  is due to the change in the lattice vibrations, it may be expected that the ratio between the shape-change effect and that of the volume-change effect (e.g.  $(\alpha_{11} - \alpha_{12})/(\alpha_{11} + 2\alpha_{12})$ ) is much larger for the thermo e.m.f. than for the resistance (by a factor 5.4 and 2.2 for Cu and Ag respectively). For gold we now find an interesting exception: The difference ( $\beta_{11} - \beta_{12} = 0.6$ ) is smaller than for copper and silver by a factor 10. According to fig. 4 this indicates that gold touches the Brillouin zone boundary with its Fermi surface in the (111) direction. The value of  $2\alpha_{44}$  for copper being rather small, we may conclude that copper certainly does not touch the zone boundary. Silver with its larger  $2\alpha_{44}$ , and still rather large ( $\beta_{11} - \beta_{12}$ ) may be an intermediate case. Hence copper has the smallest energy gaps, and gold the largest. This corresponds to what one might expect from their sequence in the periodic table, for large nuclear charges give deep potential troughs. In fig. 3 and 4 we plotted the presumable points for Cu, Ag and Au, but this has only qualitative value. In order of magnitude the computed effects agree with the effects observed, better so for the thermo e.m.f. than for the resistance effect, probably due to the closer approximation.

The considerations of Peierls<sup>8)</sup> on the "Umklappprocessen" in the theory of conductivity led to the result that most of the monovalent metals should touch the zone boundary with their Fermi surface. This point has been greatly clarified by the recent paper of Klemens<sup>9)</sup>, who showed that Peierls' calculation does not hold and that the problem of touching is still an open question. Our result for gold agrees with the conclusion arrived at by Kohler<sup>10)</sup>, who considered the magneto-resistance in high fields; this is another way of making the resistance anisotropic. Kohler showed that this magneto-resistance should be isotropic if the Fermi surface does not touch the Brillouin zone boundary and anisotropic if it does touch. Experiments<sup>11)</sup> showed that for gold it is highly anisotropic.

Approved For Release 1999/09/24 : CIA-RDP83-00423R002000130009-6

596 INFLUENCE OF SHEAR ON CONDUCTIVITY OF CUBIC METALS

For copper and silver no experimental values are available.

We have seen that electrons in the second Brillouin zone have a negative ( $a_{11} - a_{12}$ ) or  $2a_{44}$  and this can perhaps account qualitatively for the experimental values of Al. Also the negative value of ( $\beta_{11} - \beta_{12}$ ) bears this out. The fact that  $a_{11} - a_{12} < 0$  indicates that there is also an overlap of electrons in the second zone in the (100) direction. Leigh<sup>12)</sup> in his work on the elastic constants of Al arrives at the same conclusion.

This work was started at the Technische Hogeschool at Delft under the auspices of Professor M. J. Druyvesteyn, to whom the author wishes to express his indebtedness.

Eindhoven, 7 April 1952.

Received 22-4-52.

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